Response to Referee # 1:

Interactive comment on "Wet and dry deposition of atmospheric nitrogen at ten sites in Northern China" by Y. P. Pan et al.

Anonymous Referee #1

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General Comments

Although ten sites are far from enough to make clear the distribution of nitrogen deposition in Northern China, a very large area, the monitoring has been the most detail one in China till now, especially including dry deposition. Of course some uncertainty exists, however, the results, which show extremely high values, are of great importance.

Monitoring methodology is most important and need be described in detail. More information on emission inventory and site condition is necessary for analysis.

Response: The authors appreciate the valuable suggestions given by Referee # 1 for improving the overall quality of the manuscript. In the revised version we have made substantial changes by adding the uncertainty analysis and other discussions to strengthen the paper. We have also clarified the monitoring methodology and site information in the study. Detailed responses to comments are given below.

Specific Comments

P758, L8: Detail information on monitoring site, such as landuse type and local emission sources, is important and need included in the manuscript. In addition, it seems that the monitoring is not focus on forest and grassland areas. However, the effects of nitrogen such as acidification and eutrophication are mainly related to natural ecosystems. Vegetation has also great effects on dry deposition. For forest and grassland, dry deposition can be easily measured by throughfall method.

Response: In order to discuss the relevant results in a more explicit way, we have added a table in the revised paper that lists available information on monitoring site, such as meteorological parameters, land use type and local emissions.

We definitely agree with the statement that "the effects of nitrogen such as acidification and eutrophication are mainly related to natural ecosystems", however, the magnitude and potential impacts of atmospheric N deposition in China remain uncertain because of a paucity of quantitative knowledge. Numerical model is a useful tool to address these concerns, but a major problem of using this method is that observed data to validate the model is often lacking, thereby hampering accurate estimation of N deposition in the target area. Northern China is considered to be one of the main "hot reference spot" of atmospheric pollution in East Asia due to the rapid development. Therefore, ten monitoring sites were selected with varying urban geographies, energy structures and ecosystem types to provide regional information on N deposition in this region. Although the present investigation is far from clearing up all aspects of N deposition in Northern China, it adds substantially to the existing knowledge about the character and amount of the atmospheric N deposition to the surrounding environment. Most important, the present observations would be very useful to better constrain the emission inventory and atmospheric chemistry models in this part of the world. In future research, we will use models (e.g., CMAQ) to address the variations of N deposition in various areas, not only in urban, rural and agricultural regions but also in forest, grassland and coastal waterbody in China.

Vegetation has great effects on dry deposition and throughfall method was usually applied to provide an estimate of the total deposition to forest soils, including dry deposition, by measuring the amount and composition of rain water passing through a forest canopy. However, several problems were also related to this approach as summarized by previous studies (Draaijers et al., 1996; Balestrini et al., 2007). Additionally, this method was confined to ecosystems with vegetation (e.g., forest and grass) and not suitable for urban surfaces and water bodies where vegetation is lacking. Alternatively, surrogate surface and inferential method were used to estimate the particulate and gaseous dry deposition of N in this study, respectively.

P758, L25: How water examples delivered and stored before analysis need be introduced. How unstable NO2- can be well kept?

Response: Samples were frozen at each site immediately after collection. They were transported in iceboxes to the analytical Lab in Beijing and stored in a refrigerator at -20^{o}C until analysis. We have added more sentences here to clarify pre-treatment of precipitation (rainwater and snow) samples in this section.

NO_2^- is a relatively unstable form of N that is readily converted to NO_3^- and vice versa; thus, it cannot be well kept prior to analysis. The concentration of NO_2^- in environmental media is usually very low, even the concentration of NO_3^- is high. In order to investigate the inorganic N pool in precipitation and dry deposited particles, individual N species including NO_2^- were determined using ion chromatograph. The results showed that the NO_2^- is an insignificant fraction of N budget.

P759, L16: Since PM was analyzed monthly, how to reduce evaporation of NH4+ and transformation of other unstable component? The method was referred to Pan (2010b), which is however in Chinese and not easily read by international readers.

Response: To date, there is still no generally acceptable technology for sampling and analyzing particulate dry deposition flux of N ($_p$IN). Therefore, there have been insufficient data to reliably estimate and understand particulate dry deposition process. Previous studies have found that a bucket collected more dry-deposited material than Teflon, foil or coated foil surfaces (Dasch, 1985). Recently, a PUF filter based surrogate surface was placed in the glass bucket to successfully collect the dry-deposited airborne particles for chemical analysis (Pan et al., 2010). The method can avoid particle bounce and is relatively inexpensive and simple; it can be used at a variety of locations and over varying time intervals to delineate spatial and temporal information. The method was used in this study to estimate $_p$IN. We have added a detailed description of the sampling method in the revised manuscript.

For large particles, gravitational settling is a major component that can be sampled with this method. Studies have shown, however, it is difficult to capture fine particles

because the impaction and interception of particles are important for vegetative canopies and their effects are not reproduced in the design of any standardized artificial collection device (Wesely and Hicks, 2000). Therefore, the estimation of the dry deposited particles in this study has potential uncertainties and may provide an estimation of the lower limit for $_{p}$IN.

Additionally, measuring particle NH_4NO_3 is known to be plagued with problems due to its semi-volatility (Lestari et al., 2003). In previous studies, the concentrations of particulate NO_3^- collected by the weekly filter-pack was 7% lower than those by the annular denuder system (Sickles et al., 1999). Even the two existing North American inferential dry deposition networks, CASTNET and CAPMoN, have known measurement uncertainties due to the volatilization of particle NH_4NO_3, and large uncertainties in the flux estimates due to differences in their respective inferential models (Dentener et al., 2009). Since the dry deposited particles in this study were measured at ambient temperatures for a relative long sampling period, artifacts cannot be prevented due to volatilization of some reactive N species, especially NH_4^+ and NO_3^- in summer, and dry deposited N could probably be underestimated. These discussions are added in a new section to clarify the uncertainties in the measurement.

P760, L15: The dry deposition velocity is of great importance for the results. Here the data were based on modeling results, which is actually lack of test in China. I suggest adding uncertainty analysis in the discussion part.

Response: The suggestion is implemented. In this study, wet deposition and particulate dry deposition of N were collected using a custom wet-dry automatic collector at ten sites. However, measurements of gas dry deposition are technically challenging and expensive, so modeled deposition velocities are paired with measured concentrations to determine fluxes (Schwede et al., 2011). This technique has been used extensively as an operational tool to compensate for the absence of measured dry deposition data at regional scales (Flechard et al., 2010) and to estimate the total deposition flux of N for monitoring networks in Europe (EMEP), North America (CASTNET) and East Asia (EANET) (Endo et al., 2010).

The dry deposition velocity (Vd) implemented by CMAQ v4.6 in its second dry deposition scheme (M3Dry) is parameterized here following the well known resistance approach (Wesely, 1989). However, all models entail a degree of uncertainty and this study is no exception. Modeled dry deposition fluxes are very uncertain, and can seldom be compared to measurements, due to many factors including incompleteness of observations, uncertainties in chemistry schemes, and sparseness of measurements (Pleim and Ran, 2011). For example, NH_3 exhibit distinctly bi-directional behavior where it alternately deposit and emit on diurnal, seasonal, or sometimes longer time-scales, yet CMAQ v4.6 is not able to model this real-world phenomenon. Additionally, the current model works with outdated land cover databases because land cover in recent years was not available. Furthermore, the underlying surface parameters (e.g., albedo and surface roughness length) used in CMAQ was based on the measurements in European and U.S. and may different from China, possibly leading to large inconsistencies.

Although it is important to understand the dominance of these factors on the Vd variations, it still remains unclear how accurate the model is due to a paucity of measurements. Even for ozone dry deposition, which is the most widely measured substance, current model results still show substantial errors compared to observations (Pleim and Ran, 2011). Since the experimental data to confirm the calculated resistances for the large areas meant to be addressed are not available, uncertainties are difficult to quantify (Endo et al., 2010; Wesely, 1989). In previous comparison of models, relative uncertainties for weekly estimates of Vd computed of atmospheric gases for selected areas in the eastern U.S. are approximately ±30% (Wesely, 1989). Uncertainties in Vd modeling can be reduced if field measurements provide constraints on parameterizations and that is beyond the scope of this paper. To refine the quantities of dry deposition flux, follow-on field experiment will focus on comparisons of the models against NH_3 flux measured by eddy covariance.

P760, L21: Considering the seasonal pattern of deposition, paired t-test may be better than nonparametric tests for temporal and partial differences.

Response: Atmospheric deposition flux of N species was not always normally distributed, either among the ten sites or in different seasons. Therefore, differences among sites and seasons were tested with nonparametric test.

Following the suggestion, paired t-test was also conducted to examine the significance of differences throughout the manuscript and the results are consistent with nonparametric test.

P761, L15: It showed no significant difference among the sites. How is it with paired t-test?

Response: The suggestion is implemented. Please see our above response.

P762, L21: Here higher NH4+ deposition in winter was attributed to heating. I do not think heating is a major source of NH3. Rain wash may be the factor, same as for low NH4+ deposition in summer.

Response: We agree with this comment. We have modified this sentence in the revised manuscript.

P762, L24: It should be explained why NO3- deposition was high in summer and fall, maybe relating to seasonal change of NOx emission?

Response: The particulate dry deposition flux of NO_3^- ($_pNO_3^-$) showed weaker seasonal variation with a lower flux in winter than in other seasons. Compared with $_pNO_3^-$, the ambient concentrations of NO_x (Tang et al., 2012) and NO_3^- in $PM_{2.5}$ in the target areas presented a different seasonal pattern, with the high levels appearing in winter (Zhang et al., 2012; Wang et al., 2005). However, the majority of the dry deposited NO_3^- is due to coarse particles (Holsen and Noll, 1992; Lestari et al., 2003). Considering another fact that a seasonal cycle of deposition velocity of coarse particles was not evident in the time series (Nho-Kim et al., 2004), the seasonal variations of $_pNO_3^-$ is expected to be highly correlated to the concentration of NO_3^- in coarse particulate matter. This hypothesis is supported by our previous size-resolved compositional analysis that the concentration of NO_3^- in coarse particle in summer

was higher than that in winter. Both temperature and relative humidity had great impact on the size distribution of NO_3^- (Guo et al., 2010), and these factors may favor the formation of coarse mode NO_3^- in warm months, resulting in the peak values of $_pNO_3^-$ in summer and fall.

The inventory of air pollutant emissions in China in the year 2006 suggested the total NO_x emissions have weaker seasonal variations, with ratio of 1.3 between monthly emissions in December (maximum) to emissions in April (minimum), because they mainly come from industrial and transportation emissions that have less of a seasonal cycle (Zhang et al., 2009). The seasonal mismatch between emissions and $_pNO_3^-$ might be attributable to some sources that were not included in their inventory. For example, the emissions of NO_x in China usually have two peaks in May to June and October after wheat and food crops harvests, respectively, associated with the increase of biomass burning, fertilizer and other agricultural activities (Zhang et al., 2012). In addition, summer increase of NO_x emissions due to new power plants could be also a contributor for the seasonal change of $_pNO_3^-$ (Wang et al., 2012). On the other hand, the lower $_pNO_3^-$ in winter than other seasons can be partially supported by the fact that NO_x emissions in February are lower than in neighboring months, because of reduced industrial activity during the Chinese Spring Festival holiday (Zhang et al., 2009).

The above discussion has been added in the revised paper.

P763, L17: I was very surprised that no yearly difference of NO2 dry deposition existed. As I know, emission of NOx in China increased in during 2006-2010, mainly caused by increasing consumption of coal.

Response: Thanks for this insightful comment. In fact, the gaseous dry deposition flux of N ($_gIN$) was different from year to year at each site. However, according to the statistical results, the inter-annual variations for $_gIN$ was not significant at most of the sites with the exception of XL, which showed significantly lower values in 2008 than in 2009 or 2010 ($p<0.05$). Paired t-test was also performed for $_gNO_2$ and $_gNH_3$ separately, and the results were similar to that of $_gIN$.

Although differences between the three years are not significant at the 0.05 level, it is important to point out that $_gNO_2$ appear to be slightly increased year by year except for TJ and TS. The overall average $_gNO_2$ in 2010 was 22% higher than 2008. On the other hand, the trend in $_gNH_3$ is more ambiguous than that of $_gNO_2$ at most of the sites, yet the $_gNH_3$ also showed a higher value in 2010 than 2008 (8%).

Although interpreting the relationship between emissions and deposition of N without long-term measurements is challenging, our finding is consistent with the increasing trend of NO_x and NH_3 emissions in recent years due to increasing fossil fuel combustion and agricultural activities, respectively (Zhao et al., 2008).

P763, L24: The molar ratio of NH4+ to NO3- is too high. Is it reasonable?

Response: Thanks for this comment. We recognize the molar ratio of wet deposition flux of NH_4^+ to NO_3^- is expected to be similar to that of the concentrations of NH_4^+ to NO_3^-, scaling only with the total precipitation. However, the molar ratio of $_gNH_3$ to $_gNO_2$ was determined by the concentrations and Vd. Although the measured concentrations of gaseous NH_3 and NO_2 were comparable, the flux estimates were significantly different because of differences in the model-estimated Vd. Therefore, it is not suitable using the molar ratio of $_gNH_3$ to $_gNO_2$ to discuss the source information. Instead, we now use the molar ratio of gaseous concentrations of NH_3 to NO_2 in the revised paper to investigate the relative contribution from different sources.

P765, L8: The conclusion of no significant year-to-year variations is suspicious.

Response: As mentioned above, $_gIN$ values were different from year to year at each site and there was a slight increasing trend for dry deposition from 2008 to 2010, yet the year-to-year variations of wet deposition flux of N showed no trend, leading to the inter-annual variations of total N deposition more ambiguous.

We have modified the text to weaken the statement here since the year-to-year variation of N deposition is small. We have also added more discussions about this issue in the revised paper.

P765, L15: Arithmetic average value for the whole region is not good enough since the sites represent different area. Suggest giving average values to different landuse types such as urban, rural, and forest, and compare with other studies type by type.

Response: We have re-organized this paragraph and added a standard deviation for the mean value for the whole region. We also present measured values in different land use types. Note that this paragraph was aimed to compare the total N deposition with previous studies in Northern China. However, it is difficult to compare with other studies type by type (except for agriculture sites) due to the fact that the measurement data in Northern China is lacking, as summarized in the Introduction.

We believe the landuse-area weighted value is better than the arithmetic average flux for the whole region. Except for the land surfaces, however, N deposition in Northern China is also influenced by the emissions and meteorology in different areas. All of these factors will be considered in future modeling work.

P769, L4: In China, large part of NOx comes from coal combustion, the same source of SO2, which has mostly from coal combustion. I suggest adding results on sulfur deposition (at least SO4 in wet deposition), if possible, to show the contribution of different sources.

Response: Thanks for this insightful comment. A preliminary inspection of scatter plots of SO_4^{2-} versus NO_3^- revealed that relationships were reasonably well explained by linear models at each site ($0.56< r^2 <0.94$, $p<0.001$), indicating some similarities in the sources and sinks of these species (Arimoto et al., 1996). This can be due to the fact that in China large part of NO_3^- comes from coal combustion, which is the major source of SO_4^{2-} (Zhang et al., 2009). However, the sources for atmospheric NO_3^- are considerably more diverse than for SO_4^{2-}, especially in urban areas where the contribution from vehicle emission cannot be ignored (Huebert et al., 1988). Following previous studies (Huebert et al., 1988; Arimoto et al., 1996; Yao et al., 2002), the mass ratio of NO_3^- to SO_4^{2-} was used as an indicator to investigate the relative importance of stationary versus mobile sources of sulfur and nitrogen in wet deposition. We have added more discussions about this issue in the revised paper.

PP770, L2: The contribution of NH4+ is less than NO3- in PM deposition!? Is it due to NH3 evaporation? Result here does not coincide with many studies on component of PM in this area.

Response: It has been found in previous studies that the contribution of NO_3^- to $PM_{2.5}$ and PM_{10} is larger than that of NH_4^+, whereas in terms of N, the concentration of NH_4^+-N is larger than NO_3^--N in both $PM_{2.5}$ and PM_{10} (Wang et al., 2005; He et al., 2001). However, large particles play a significant role in dry deposition flux because of their deposition velocities. For example, it was found that the majority of the total fluxes for particulate mass and both SO_4^{2-} and NO_3^- is due to coarse particle (Holsen and Noll, 1992; Lestari et al., 2003). Therefore, the composition of dry deposited particle is expected to be similar to the coarse particulate matter. Evidence is our previous size-resolved compositional analysis performed for particles between 0.01 and 100 μm diameter obtained from field measurements with a cascade impactor in winter. It was found at the BJ, TJ and TS sites that the concentration of NH_4^+-N is less than NO_3^--N in coarse particle with diameter larger than 2.1 μm; however, at the BD and XL sites the reverse is true (Sun et al., 2011). This finding coincides with our studies on component of dry deposited particles.

It should be noted that NH_4^+-N played a greater role than NO_3^--N at most of sites with the exception of BJ, TJ, TS and YF. At these 4 sites the contribution of NH_4^+-N is less than NO_3^--N in particulate N dry deposition. This cannot be attributed to NH_3 evaporation because the volatilization of NH_4^+ in this study could be a systematic error for all the ten sites and not limited to a few sites. According to the above discussions, we suggest that the relative higher molar ratio of NH_4^+/NO_3^- at these 4 sites was attributed to the enhanced contribution of NO_3^-. This result is not surprising because these industrial and urban areas are strongly affected by NO_x emissions from consumption of fossil fuels. This can be further supported by the measurements that NO_3^- had a larger coarse mode in polluted urban areas of Beijing than its upwind rural site (Guo et al., 2010).

In the revised version, we have added above discussions.

P771, L1: Critical loads are evaluated for vegetation. For forest and grassland in this area, comparison of critical load to deposition derived in urban and agricultural area is not reasonable. The deposition in forest and grassland areas may be much lower than the results got here, it may thus overestimate the effects of nitrogen impacts.

Response: We agree with this insightful comment that comparison of deposition data derived from vast areas (including urban and agricultural sites) to critical loads used for forest and grassland areas is not reasonable. Since the XL site is located in forest areas, comparison of critical loads to N deposition derived from this site is used in the revised paper to primarily evaluate the effects of N in such land use type.

P772, L13: The reference of Tang (2010) may be not available for most readers. Can you refer to some other studies?

Response: A more in-depth description and discussion of the emission inventory in the target areas is included in another manuscript titled “high-resolution ammonia emissions in Northern China, 2006-2011”. The manuscript is in preparation by G. Tang et al., and will be submitted to an international journal in the near future.

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Response to Referee # 2:

Interactive comment on "Wet and dry deposition of atmospheric nitrogen at ten sites in Northern China" by Y. P. Pan et al.

Anonymous Referee #2

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General comments:

The topic of the paper is about wet and dry deposition of oxidized and reduced nitrogen compounds in Northern China, using a three-year data base. Deposition amounts are high, and measurements have been made in sites dominated by anthropogenic emissions such as fertilizers, power plants, and traffic. The paper is well written and easy to read, except the last part before the conclusion which is a bit confusing. The paper gives novel and interesting results on N deposition in a region where emissions are supposed to increase in the near future. Revisions are needed before publication.

Response: The authors appreciate the valuable suggestions given by Referee # 2 for improving the overall quality of the manuscript. In the revised version we have made significant changes by adding the discussions of gases concentrations and deposition velocities to strengthen the manuscript. We have also clarified the site conditions and modeling of the deposition velocity in the study. The last part before the conclusion has been re-organized to make it easier to follow by readers. Please refer to our response for more details given below.

However, several points can be addressed to improve the quality of the paper:

- The description of the sites needs to be done better than giving a reference. The reader needs to know more precisely what type of emissions will influence the deposition fluxes at the different sites. Some explanations are given at the end of the paper, but this arrives far too late. A table should be included with site description in

terms of mean annual meteorological parameters (rainfall, temperature), local emissions, density of population, soil type, and other relevant information specified in the specific comments.

Response: As suggested, a description table is added in the revised paper that lists available information on monitoring sites, such as meteorological parameters, land use type and local emissions. Now the relevant results can be discussed in a more detailed way.

- The modeling of deposition velocities needs to be better described. What parameterizations are used? Is the compensation point concept used for the calculation of NH3 deposition velocity? At what height are calculated the velocities? Is there a seasonal variation in these velocities for each specific compound?

Response: Thanks for these comments and suggestions. We have added the following descriptions to make this section more clearly.

The default dry deposition velocity (Vd) implemented by CMAQ v4.6 in its second dry deposition scheme (M3Dry) is parameterized following the well known resistance approach (Wesely, 1989), which is computed coherently with MM5 land-surface model. Three resistances are considered in this theory: aerodynamic resistance (Ra), quasi-laminar resistance (Rb), and surface or canopy resistance (Rc), among which the canopy resistance is the most difficult one to simulate (Zhang et al., 2008). The Vd is expressed as:

$Vd=(Ra+Rb+Rc)^{-1}$

Vd used in the present study was taken from a height of 38 m, which is the centre of the lowest layer in CMAQ model. There is seasonality in Vd for each N species. We have added a new section to discuss the variation of Vd in the revised paper.

Different from most gases that are consistently deposited, the surface-atmosphere exchange of NH_3 is bi-directional, but in CAMQ v4.6 it is treated as dry deposition only (Byun and Ching, 1999). Since the evaporation of the deposited NH_3 from surface was not taken into account in the current version of CMAQ model, the flux calculated in the present study represents a rather non-conservative N deposition

estimate (upper boundary). The overestimation may be significant over managed agricultural ecosystems because the canopy compensation point is generally larger over agricultural crops and fertilized vegetation (Zhang et al., 2010). Therefore, the effects of the stomatal compensation point on NH_3 deposition in agricultural sites due to the counterbalance between deposition and emission are discussed in the revised paper. As reduced N becomes a larger fraction of the reactive N budget, wide-spread NH_3 measurements and improved NH_3 Vd modeling are critical needs in future.

- Monthly means for deposition velocities and concentrations are needed for comparison with other studies.

Response: The suggestion is implemented. In the revised paper, we have added a new section to discuss the variation of monthly Vd and concentrations of N species. We have also compared the observed concentrations and modeled Vd with other studies.

- The interannual variation is not significant, according to the authors. But why? This point should be addressed in more details.

Response: This issue has been addressed in the response to Reviewer 1 (P763, L17 and P765, L8). We have modified the text to weaken the statement here since the year-to-year variation of N deposition is small. In brief, it is true that the observed gas dry deposition flux of N ($_gIN$) were different from year to year at each site. However, the inter-annual variation for gIN was not significant at most of the sites according to the statistical test. Although differences between the three years are not significant at the 0.05 level, the $_gNO_2$ appear to be slightly increased year by year at most of the sites. Our finding is consistent with the increasing trend of NO_x emissions in recent years due to increasing fossil fuel combustion (Zhao et al., 2008).

Although there was a slight increasing trend for dry deposition from 2008 to 2010, the year-to-year variations of wet deposition of N showed no trend, leading to the inter-annual variations of total N deposition more ambiguous. More discussions about this issue were added in the revised paper.

- Uncertainties should be given for wet and dry deposition fluxes, as well as for emission estimates.

Response: Wet deposition of N could be underestimated due to light rain samples were not included in the dataset. On the other hand, evaporation of NH_4^+ and NO_3^- during the sampling period may result in the underestimation of particulate dry deposition of N. Uncertainties also existed in the gas dry deposition because the Vd is based on modeling results, which is lack of test. Therefore, we added a separate section to discuss the uncertainties in estimating wet and dry deposition fluxes in the revised paper based on the suggestion.

As for the uncertainties in emission estimates, it will be included in another manuscript titled "high-resolution ammonia emissions in Northern China, 2006-2011", which will be submitted to an international journal in the near future. This would help focus the manuscript on the N deposition.

Specific comments

P757 L8: "for another purpose": what purpose?

Response: Another purpose here means that these measurements were not aimed to study N deposition. To clarify the statement, the sentence has been rewritten as: Although N species in gaseous, particulate and rain phases have been measured in various locations in Northern China, relatively few studies have primarily focused on N deposition measurement (Meng et al., 2011; Zhang et al., 2007; Tang et al., 2005).

P758 L24: Describe in a few words the procedure before referring to Pan et al. (2010a)

Response: The suggestion is implemented. We have added more sentences here to describe the procedure handing the rainwater and snow samples.

P759 L 5-8: The volume-weighted concentrations and the amount precipitation are multiplied to obtain the wet deposition flux. But what is the time scale of the integration: week? month?

Response: In the present study, daily precipitation samples were collected and analyzed. The precipitation amount was measured by a standard rain gauge at each site. To calculate the monthly, seasonal and yearly wet deposition flux of N, the volume-weighted concentrations of N species and the amount of precipitation during the corresponding period are multiplied.

P759 L13: Pan et al. 2010b is in Chinese. Is it possible to have another reference in English?

Response: Alternative reference is not available, but in the revised paper, the sampling and analysis method is described in detail for readers' convenience.

P760 L3: Same question as for wet deposition fluxes: what is the time integration?

Response: The ambient NO_2, NO_x and NH_3 concentrations were measured monthly using the Analyst diffusive cubes exposed simultaneously for a month. Based on the monthly concentration data and the modeled Vd of each compound during the corresponding period, seasonal and yearly dry deposition flux were integrated.

P760 L7: What is the height of measurements?

Response: The ambient concentrations of gaseous N were measured at the similar height to that of the automatic wet-dry sampler. At most of the sites, the measurements height is about 1.5 m above the ground. On the other hand, at the BD, BJ, CZ, TJ and TS sites, the sampler was installed at the rooftop or tower with a height varied from 3 to 10 m above the ground. This detailed information is included in the site description table in the revised version.

P760 L15-19: As mentioned in the general comments, this paragraph is too short and does not give sufficient explanation on how the deposition velocities are calculated. What is the height chosen for the calculation in the model? Is it in accordance with the height of measurements? If not is there a correction made? Deposition velocities can be quite different depending on the wind velocity (refer to Zhang et

al.,doi:10.1029/2008JD010640, JGR, 2009). A range of variation of deposition velocities used in this study could be useful, as well as a range of variation of concentrations, for each compound taken into account.

Response: Thanks for these suggestions and comments. This paragraph is re-organized to include a more in-depth description on how the dry deposition velocities are calculated using the CMAQ model. A brief description of the height chosen for calculation and measurements is provided in our above response. Although Vd is not calculated according to the height of measurements, it results in insignificant errors in dry deposition flux estimates because the differences of Vd between 10 and 50 m are estimated to be only a few percent under neutral and unstable conditions in previous studies (Zhang et al., 2005). On the other hand, during the night when the atmospheric conditions are stable, Vd were lower due to the limited vertical dispersion of pollutants (Zhou et al., 2010).

Since the Vd is simulated by CMAQ, the influences of wind velocity on the deposition velocities have been considered in the meteorology field derived from MM5. In addition to the fluxes, two paragraphs are added in the revised paper to discuss the variations of gaseous concentrations and Vd of N species.

P761 L19: In Fig 3a-b, could you add the monthly rainfall?

Response: We accepted the suggestion and have added a new figure showing the monthly mean precipitation amount at ten sites investigated.

P762 L4: A reference to a table containing site information is needed here. Is it possible to have an idea of the amount of fertilizers used in each agricultural site? Is there a possible influence of long range transport of pollutants such as NH3 to explain such high deposition fluxes, or local sources are sufficient to justify these amounts?

Response: We have added the fertilization information and related discussion in the revised paper to address above points suggested by the referee.

Farmers typically apply about 500-600 kg N ha^{-1} yr^{-1} as N fertilizer to achieve high yields of maize and wheat in this region. Of the applied N fertilizer, however, less

than 30% will be absorbed by the crops and more than 20% (ca. 100 kg N ha^{-1} yr^{-1}) is lost by NH_3 emission (Shen et al., 2009). This makes a significant contribution to high deposition fluxes in agricultural regions. However, the source of NH_3 at a given site is expected to be influenced by a broader area than just local emissions.

P762 L13: Site's rural characteristics: more explanation is needed (cf site description table)

Response: More information of monitoring sites is added in a description table. Of the ten sites, XL is located in mountain areas and surrounded by forest and few villages, it represents rural characteristics in the target areas. Since there are no serious local emissions within 100 km, it was also used as a background station to understand the regional air pollution in Northern China (Xin et al., 2010).

P762 L26: Why pNO3- is notably higher in July August and September at the BJ site?

Response: This issue has been addressed in the response to Reviewer 1 (P762, L24). Because the majority of the dry deposited NO_3^- is due to coarse particles (Holsen and Noll, 1992; Lestari et al., 2003) and the seasonal cycle of deposition velocities of particles is nearly undetectable (Nho-Kim et al., 2004), the seasonal variations of $_pNO_3^-$ is related to the concentrations of NO_3^- in the coarse particles. This is supported by our previous size-resolved compositional finding that the concentration of NO_3^- in coarse particle in summer was higher than that in winter.

Both temperature and relative humidity had great impacts on the size distribution of NO_3^- (Guo et al., 2010), and these factors may favor the formation of coarse mode NO_3^- in warm months. Therefore, $_pNO_3^-$ is larger in summer than winter at most sites. Detailed discussion about the seasonal variation of $_pNO_3^-$ is added in the text.

P765 L15: Give a standard deviation for IN deposition flux.

Response: A standard deviation for IN deposition flux was added here. This paragraph was re-organized as suggested by reviewer 1 (P765, L15).

P768 L3: Do you mean dissolved organic nitrogen?

Response: To support the fact that the rainfall amount was an important controlling factor on the seasonal trends of wet deposition, here using the relationship between the dissolved organic carbon and precipitation amount as an example (Pan et al., 2010).

P768 L15: What is the scavenging ratio? Why do you assume it is constant at all sites?

Response: We have clarified the concept in the revised manuscript. The concept of scavenging ratios is based on the simplified assumption that the concentration of a component in precipitation (C_p) is related to the concentration of the respective compound in the air (C_a). Thus scavenging ratio can be calculated on a mass basis:

$W=C_p/C_a$

When the precipitation amount is expressed as P, the wet deposition flux of the constituents (F) depends on W, C_a and P by:

$F= WC_aP$

Therefore, if the scavenging ratio and atmospheric concentrations are constant at all the sites, the wet deposition flux increase in proportion to the precipitation amount. However, for the sites with higher concentrations of pollutants, the wet deposition flux was greater than those expected from the amount of precipitation based on the above premise (i.e., WC_a=constant). This allows us to evaluate the degree influenced by anthropogenic emissions at each site, using the relationship between the wet deposition of measured species and precipitation amount (Sakata et al., 2006). These descriptions are added in the revised manuscript to make this section easier to follow.

P770 L19: Information about the timing of fertilization should be given in the site description table.

Response: The suggestion is implemented.

N fertilizer is usually applied to fields in March-April, July-August and October each year. More fertilization information was added in site description table.

P770 L23: This title is not appropriate in my opinion.

Response: We accept this suggestion and have modified the title. This section is re-organized in the revised paper.

P771 L16-end of paragraph: This paragraph is a bit confusing and it is difficult to understand exactly what has been done by the authors. Is it a comparison between several simulations, a specific simulation performed for this study, results taken from other studies? Several sub paragraphs are needed to clarify.

Response: Thanks for this comment. In the revised version, these paragraphs are re-organized and clarified to avoid confusion.

P773 L26: Forests and grasslands are not part of the measurement sites, and deposition fluxes on this type of ecosystems should be lower than in urban or agricultural sites.

Response: We agree with this comment and this sentence is rewritten. To primarily evaluate the effects of N deposition in natural ecosystems, in the revised paper we compared the critical loads for temperate forests to N deposition derived from the XL site, which is located in forest areas and surrounded by few villages.

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